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Nitrous oxide emissions from China's croplands based on regional and crop-specific emission factors deviate from IPCC 2006 estimates

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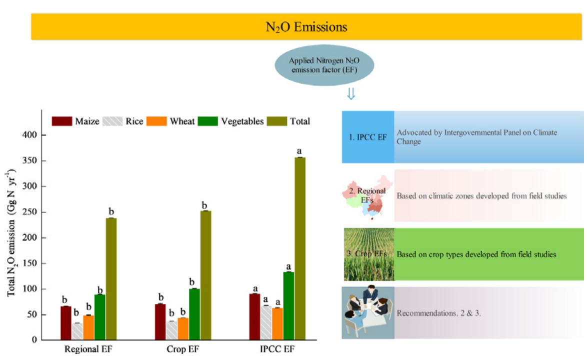
HIGHLIGHTS

- Croplands in China had a mean soil nitrous oxide emission factor of 0.60%.
- This emission factor (EF) from China's croplands varied among climatic zones.
- Precipitation and soil pH contributed significantly for regional EFs variation.
- IPCC's default EF would overestimates N_2O emissions from China's croplands.
- The relationship of EF to soil and climatic variables was non-linear (quadratic).

GRAPHICAL ABSTRACT



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ABSTRACT

Calculated N_2O emission factors (EFs) of applied nitrogen (N) fertilizer are currently based upon a single, universal value advocated by the IPCC (Inter-governmental Panel on Climate Change) even though EFs are thought to vary with climate and soil types. Here, we compiled and analyzed 151 N_2O EF values from agricultural fields across China. The EF of synthetic N applied to these croplands was 0.60%, on average, but differed significantly among six climatic zones across the country, with the highest EF found in the north subtropical zone for upland fields (0.93%) and the lowest in the middle subtropical zone for paddy fields (0.20%). Precipitation and soil pH, which showed non-linear relationships with EF, are among the factors governing it, explaining 7.0% and 8.0% of the regional variation in EFs, respectively. Annual precipitation was the key factor regulating N_2O emissions from synthetic N fertilizers. Among crop types, legume crops had the highest EFs, which were significantly ($P <$ 0.05) higher than those of cereals. Total soil N_2O emissions from fertilized croplands with maize, rice, wheat, and vegetables in China, calculated using the climatic zone (regional) EFs, were estimated to be 239 Gg N yr^{-1} with an uncertainty of 21%. Importantly, this value was substantially (33%) lower than that (357 Gg N yr^{-1}) derived from the IPCC default EF but close to the 253 Gg N yr^{-1} estimated using crop-specific EFs. N_2O emissions from applied synthetic N fertilizer accounted for 66.5% of the total annual N_2O emissions from China's maize,

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rice, wheat and vegetable fields. Taken together, our study's results strongly suggest that regional EFs should be included for accurate N_2O inventories from croplands across China.

1. Introduction

The global atmospheric nitrous oxide (N_2O) concentration has increased to approximately 324 ppb, in 2011 (IPCC, 2013). Agricultural activities are a major contributor to rising global N_2O, particularly the addition of nitrogen (N) fertilizers, manure, and urine patches (Stehfest and Bouwman, 2006; Zaman and Nguyen, 2012). Global N fertilizer use is anticipated to increase threefold by 2050 (Tilman et al., 2002; Alexandratos and Bruinsma, 2012) to meet the expected doubling of food production required to feed the world's growing population (Mueller et al., 2012), which will likely make China one of the largest consumers of N fertilizer (Heffer, 2016). Increasing global N fertilization will accelerate the mineral N available in soil for nitrification and denitrification processes: the primary processes underpinning N_2O emissions (Senbayram et al., 2009).

Soil N_2O emissions, however, are highly heterogeneous in space and time given their association with both climatic factors and agricultural management practices (Brown et al., 2001). The relationship between N input and N_2O emission established from previous studies (Eichner, 1990; Bouwman, 1996) motivated the concept of emission factor from N applied. The emission factor (EF) is defined as a fraction of the inputted N released in the form of N_2O within the current seasonal or annual period (Lu et al., 2006), which is the N_2O emission from N fertilizer plots minus the N_2O emission from the unfertilized plots expressed as a percentage of N applied (all other conditions remained equal in all plots) (Zou et al., 2009). Yet according to the IPCC Tier 1 guidelines (IPCC, 2006), a single default EF value of 1.0% is recommended. Clearly, this overlooks the influence of regional differences in climatic type, which may well lead to inaccurate estimations of overall regional or global N_2O emissions (Lesschen et al., 2011). However, there is mounting evidence of regional N_2O EFs showing significant variation under different climatic and field conditions in China (Zou et al., 2005a, 2005b; Chen et al., 2008; Zhai et al., 2011; Wang et al., 2012; Liu et al., 2012; Shepherd et al., 2015; Niu et al., 2017), heterogeneity that was attributed to N addition and environmental variations yet was ignored in previous EF calculations (Zhou et al., 2015). Different climate types may foster different EFs, thereby fuelling controversy over the general applicability and usefulness of the IPCC guidelines (de Klein et al., 2010).

For example, the IPCC default EF value for N_2O estimation has been shown to cause relatively large discrepancies of $\pm$50% mostly due to serious limitation in observational data collected at the proper temporal and spatial scales (de Klein et al., 2006; Tang et al., 2006; Song et al., 2009; Corazza et al., 2011; Outram and Hiscock, 2012; Tian et al., 2012; Griffis et al., 2013; Zhu et al., 2013). N_2O fluxes are associated not only with the amount of N fertilizer applied but also with soil, agronomic, and climatic conditions of where it applied (Pang et al., 2009; Wang et al., 2011). In China, N_2O emission estimates for national inventories were based on averaged values from observed experimental data (Lu et al., 2006; Shepherd et al., 2015). However, due to the widely different climate types in China, regional variability in N_2O emissions is expected (Lu et al., 2006), thus creating considerable uncertainty in the overall N_2O emission estimates when relying on one default value alone (Luo et al., 2013), such as that from IPCC's 2006 methodology. Zheng et al. (2004) and Brocks et al. (2014) both reported regional variation in N_2O emissions and suggested that separate EFs for various cropland categories or regional EFs would provide more accurate estimates than those obtained by using the IPCC default EF. Similarly, Gao et al. (2011) also advised that using region-specific EFs for China would reduce some of the uncertainty associated with using the IPCC default EF (Zheng et al., 2004), provided that sufficient measurement data was available. To date, however, studies focusing on EF for specific climatic zones within a country under various cropping systems remain scarce. Currently, the N_2O EFs used in China for its national inventories are crop-specific EFs—i.e., 0.74% for upland crops, 0.75% for vegetables and orchards, and 0.30% for paddy rice—based on 60 sets of experimental data published between 1982 and 2003 (Lu et al., 2006). Since then, more field measurements have been made, deeming it both timely and necessary to propose crop-specific and region-specific EFs increase the accuracy of China's national inventory (Shepherd et al., 2015).

In this study, we compiled N_2O emissions data together with soil and environmental data measured in fields across China to propose regional climatic EFs and to evaluate the contribution of crop type to these N_2O emissions. The study's specific objectives were (i) to determine the EFs for various climatic zones and crop types in China; (ii) to assess the efficacy of the IPCC default EF value for estimating N_2O emissions in different climatic zones; and (iii) to quantify the contribution of major cropping systems to N_2O emissions in China.

2. Materials and methods

2.1. Data collection

The datasets comprised 235 N_2O field measurements sourced from peer-reviewed, English and Chinese academic journals. Data published in English were gathered from the Web of Science and those in Chinese from the China Knowledge Integrated Database (CNKI). Collectively, the experimental field studies encompass different climatic zones in China (Fig. 1). From them, the measurement values selected for use in our study adhered to the following criteria: (1) the measurements must have been made under field conditions; (2) field treatments must have been replicated at least three times; (3) N_2O emission measurements must have been taken hourly to biweekly; (4) the measurement period must have spanned at least one growing season; (5) the control treatment where no N fertilizer was applied must have been included; and (6) added N should be derived from synthetic fertilizer rather than organic fertilizer. We excluded measurements of N_2O emissions from studies using nitrification inhibitors and biochar treatments (Wang et al., 2011; Niu et al., 2017), as were any N_2O emissions from potted, incubation, and greenhouse studies.

We summarized the dataset by amending the FAO's approach, where the reported factors regulating N_2O emissions include climate and crop type, much in the same way as Shepherd et al. (2015). Our detailed database is shown in Table S1. All values of cumulative N_2O emissions were first converted to kg N_2O-N ha^{-1}; if the actual emission factor (EF) was not originally reported, we followed IPCC's, 2007 methodology to calculate the EF, by using $EF = (E_{\mathrm{N2O\text{-}fertilizer}} - E_{\mathrm{N2O\text{-}control}})$ / N applied, where $E_{\mathrm{N2O\text{-}fertilizer}}$ and $E_{\mathrm{N2O\text{-}control}}$ (kg N_2O-N ha^{-1}) are soil N_2O emissions in the N fertilizer added treatment and control (without N fertilizer) treatment, respectively; and N (kg N ha^{-1}) is the application rate of synthetic N fertilizer. That is, EF is defined as the difference between the N_2O emission from N fertilized plots minus the N_2O emission from control plot without N fertilization divided by the N rate, expressed as a percentage of the N applied.

Essential site data (sowing area) on crop field cultivation were obtained from the China Statistical Year Book (2016) and are provided in Table S2. The application rate of N fertilizer in China (Table S3) was obtained from Huang and Tang (2010) and Wang et al. (2010). The N_2O EFs of N fertilizer applied were then classified according to an

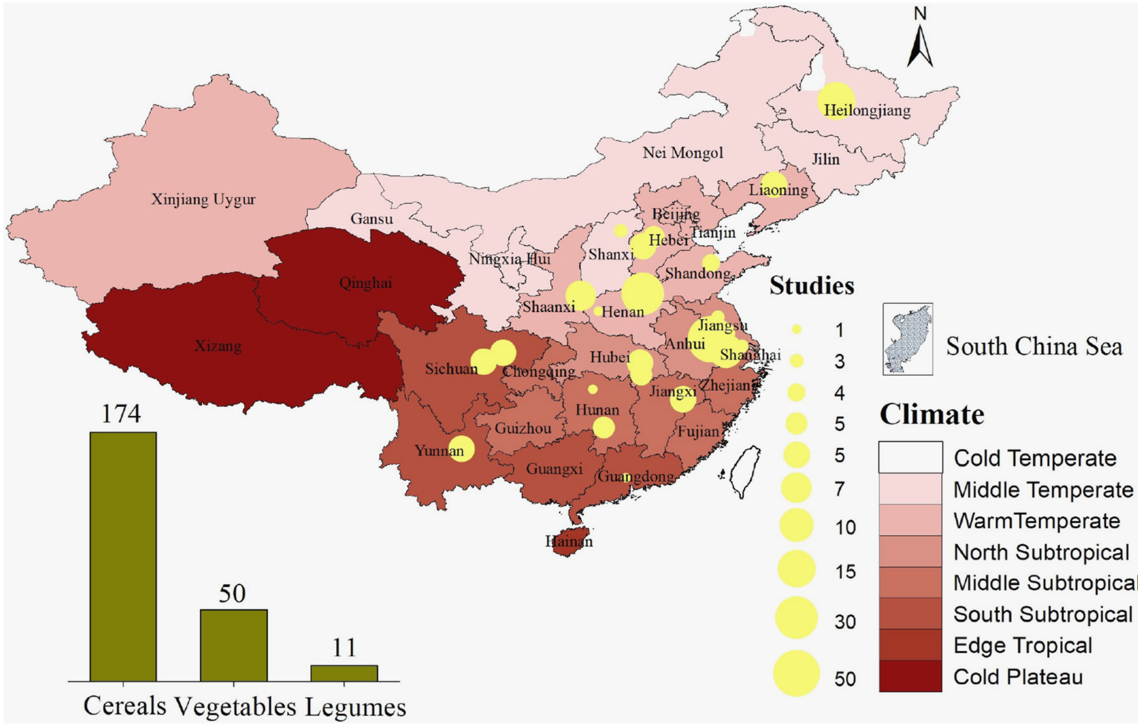

Fig. 1. Farm sizes, their locations, and the crop types used in this review study. The size of each circle is proportional to the number of N_2O measurements taken at each location.

established scheme (Zheng et al., 2010), into six main climatic zones: cool temperate ($n = 10$), middle temperate ($n = 8$), warm temperate ($n = 26$), north subtropical ($n = 80$), middle subtropical ($n = 18$) and south subtropical zone ($n = 9$) (Fig. 1). Due to a lack of field data, other climatic zones in China, such as cold plateau and edge tropical zones, could not be included in our study.

2.2. Data analysis

Out of the 235 field measurements, there were a total of 151 unique EFs as our final sample size. To quantify the relationships between N_2O emission rates and EFs with the other soil and climatic factors, we used piecewise regression function by separating different parts of the data using more than one linear model and compare with the non-linear quadratic model using same data to find the best fit. All data were statistically analyzed and log-transformed if necessary prior to analysis using SPSS v22.0 software for Windows (SPSS Inc. Chicago, USA), with one-way ANOVA followed by the least significant difference (LSD) at $P < 0.05$ used to test for significant differences among the climatic zone group means and OriginPro v8.5.0 (Origin Lab Corporation, Northampton, USA).

Based on the database analysis, two N_2O EFs of synthetic N applied were established and compared with the IPCC default EF value of 1.0%. Specifically, these three EFs corresponded to (i) a regional-scale default EF value per climatic zone for upland and paddy fields, EF–1 (known as method 1); (ii) a crop-specific default EF value applied across China, that is one single specific EF per crop used across different climate zones, EF–2 (method 2); and (iii) the IPCC default EF value (method 3). With these EFs, total N_2O emissions were then estimated from various agricultural cropping systems in China at provincial, regional, and national scales on the basis of their crop-sown area (China Statistical Year Book, 2016). However, in the climate zone where there is no enough available data for such classification, the national average EFs were used for N_2O estimation.

This classification was necessary to facilitate comparison of these two methods with IPCC constant value. Specifically, the N_2O emissions during the crop season (Y) were calculated using the following linear model equation (Akiyama et al., 2005; H. Chen et al., 2015, J. Chen et al., 2015).

$$Y = aX + b \tag{1}$$

where a is the emission factor of N applied (EF); X is the applied N fertilizer rate; and b is the soil background N_2O emission rate. N_2O emissions from synthetic N applied in crop fields ($\boldsymbol{E}_{\text{N2O-N}}$, kg N_2O-N ha^{-1}) were estimated as follows:

$$\boldsymbol{E}_{\text{N2O}-\text{N}} = EF \times N_{\text{input}} \times C_{\text{mrwv}} \tag{2}$$

where N_{input} (kg N ha^{-1}) is the application rate of synthetic N fertilizers for each crop; and C is the sowing area (ha) per crop type for maize (m), rice (r), wheat (w) and vegetables (v) under 2016 cultivation. Both sowing area (Table S2) and N application rate (Table S3) during the crop season are statistized in the provincial, regional (climate zone) and national scales. Hereafter, the term "*EF*" refers to the either regional EFs of N applied for upland and paddy fields (method 1) or crop-specific EF (method 2). In this study, we obtained one maize (wheat or vegetable)-specific EF, but paddy rice-specific EFs in different climate zones (Tables 1 and 2). In the cold and middle temperate zones, there are no rice-specific EFs for paddy field and mean rice-specific EF is used for estimation.

We used linear model here (Eq. (2)) because, for example, in China, the nonlinear models occasionally overestimated N_2O emissions by 6–15% than linear predictions from administrative units with very high N application rates, while generated underestimation bias with aggregated fertilizer data (Gerber et al., 2016). The predictions of N_2O emissions generated from the IPCC default EF method were compared with each of the confidence intervals for our two selected methods by using their lowest and upper limit range of uncertainty on predicted N_2O emissions. The most extreme percentiles obtained with the two selected methods can be interpreted as the best-case or worse-case (Philibert et al., 2012).

Table 1
Soil background N_2O emission (BNE) rates and N_2O emission factors (EF) of N applied from upland (maize, wheat and vegetable) and paddy fields in six climatic zones of China.

Climatic zone	Upland field				Paddy field			
	Number	BNE rate (kg N ha^{-1} yr^{-1})	Number	EF (%)	Number	BNE rate (kg N ha^{-1} yr^{-1})	Number	EF (%)
Cold temperate	2	0.60 ± 0.26	10	0.40 ± 0.06	–	–	–	–
Middle temperate	15	0.90 ± 0.22	8	0.64 ± 0.11	–	–	–	–
Warm temperate	13	0.61 ± 0.18	24	0.65 ± 0.10	2	0.62 ± 0.58	2	0.21 ± 0.19
North subtropical	20	1.07 ± 0.24	26	0.93 ± 0.15	21	0.46 ± 0.09	54	0.54 ± 0.06
Middle subtropical	8	0.78 ± 0.07	8	0.21 ± 0.03	7	0.55 ± 0.13	10	0.20 ± 0.05
South subtropical	5	1.55 ± 0.75	8	0.88 ± 0.17	1	0.93	1	0.84
Total	53		84		31		67	
Mean		0.92 ± 0.12		0.69 ± 0.06		0.43 ± 0.07		0.48 ± 0.05

Values are mean ± standard error. –: no data. Number: the number of measurements.

3. Results

3.1. Soil background N_2O emissions

The mean soil background N_2O emission (BNE) rate from croplands in China was 0.92 kg N ha^{-1} yr^{-1} for uplands (n = 84) and 0.43 kg N ha^{-1} yr^{-1} (n = 31) for paddy fields, with the highest (1.55 kg N ha^{-1} yr^{-1}) for uplands in the south subtropical zone and lowest (0.46 kg N ha^{-1} yr^{-1}) for paddy fields in the north subtropical zone (Table 1). Among crops, the highest BNE rate was in vegetable fields at 1.30 kg N ha^{-1} yr^{-1} (n = 19), followed by legume fields at 1.2 kg N ha^{-1} yr^{-1} (n = 6), whereas the cereal fields exhibited a significantly lower BNE rate of 0.55 kg N ha^{-1} yr^{-1} (n = 59) (Table 2). Total national BNE from maize, rice, wheat and vegetable croplands in China was estimated at 80.1 Gg N yr^{-1} when using the crop-specific BNE rates. The highest BNE we found was for vegetable fields (28.6 Gg N yr^{-1}), followed by maize (23.3 Gg N yr^{-1}), wheat (15.2 Gg N yr^{-1}) and rice (13.0 Gg N yr^{-1}) (Table S4). Using the BNE rate (b value) in Eq. (1), total national BNE from maize, rice, wheat and vegetable croplands was estimated at 85.9 Gg N yr^{-1} (Table S5).

3.2. Regional and crop-specific N_2O emission factor of N applied

The N_2O emission factor of synthetic N fertilizers applied from agricultural soils in China averaged 0.60% (range of 0.01–3.73%, n = 151), with the uncertainty confidence interval of 0.51–0.69% (Fig. 2). Among climatic zones, the highest mean EF was 0.93% for the north subtropical zone, which exceeded that of the cold temperate zone (0.40%), with the lowest EF (0.21%) found in the middle subtropical zone for upland fields (Table 1). For crop-specific EFs, the highest EF was found in legumes (soybean) at 0.90% (range of 0.10–1.70%; n = 4), followed by cereals, upland rice had the highest EF at 0.87% (range of 0.13–1.55%; n = 3), followed by maize at 0.71% (range of 0.08–1.90%; n = 22), wheat at 0.59% (range of 0.02–1.84%; n = 23), and then paddy rice, with the lowest value of 0.48% (range of 0.01–1.80%; n = 67) (Table 2). Among the vegetables, cabbage had the highest EF; others, such as lettuce and amaranthus, were grouped together due to insufficient field data per type (Table 2).

The piecewise regression analysis revealed significant non-linear relationships of EF with both precipitation (R^2 = 0.07, n = 151, P < 0.01) and soil pH (R^2 = 0.08, n = 82, P < 0.05), but not with N rate (R^2 = 0.004, n = 151, P = 0.77), SOC (R^2 = 0.01, n = 103, P = 0.54), total N (R^2 = 0.02, n = 102, P = 0.29), or air temperature (R^2 = 0.003, n = 151, P = 0.82) (Fig. 3). A significant relationship was observed between N_2O emissions as function of fertilizer N application rate (R^2 = 0.37, n = 151, P < 0.001), mean annual precipitation (R^2 = 0.03, n = 235, P < 0.05), SOC (R^2 = 0.06, n = 160, P < 0.01), and soil pH (R^2 = 0.05, n = 130, P < 0.05), but not so with air temperature (R^2 = 0.005, n = 235, P = 0.58). Finally, soil pH was significantly related to precipitation (R^2 = 0.38, n = 130, P < 0.01) and air temperature (R^2 = 0.12, n = 130, P < 0.01) (Fig. 4).

3.3. Quantifying N_2O emissions from croplands

The total N_2O emission (BNE plus N_2O emission from synthetic N fertilizer applied) from maize, rice, wheat, and vegetable fields in 2016 in China was estimated at 239 Gg N yr^{-1} when using method 1, with an uncertainty of 21% (Table 3). The highest N_2O emission came from

Table 2
Soil background N_2O emission (BNE) rate and N_2O emission factors of N applied in various crop systems in China.

Crop type	N_2O emission from N applied			Background N_2O emission	
	Number of measurements	Application rate of N (kg N ha^{-1})	Emission factor (%)	Number of measurements	Rate (kg N ha^{-1} yr^{-1})
Maize	22	75–300	0.71 ± 0.10bc	11	0.63 ± 0.09
Upland rice	3	103.5–150	0.87 ± 0.41b	3	0.38 ± 0.07
Paddy rice	67	100–450	0.48 ± 0.05d	31	0.43 ± 0.07
Wheat	23	135–850	0.59 ± 0.09bc	14	0.63 ± 0.16
Cabbage	8	180–900	0.77 ± 0.21b	4	1.9 ± 0.69
Celery	5	180–1200	0.57 ± 0.44c	3	2.6 ± 0.74
Cucumber	6	162–1200	0.28 ± 0.05e	2	0.82 ± 0.38
Rapeseed	5	150–250	0.55 ± 0.07c	2	0.3 ± 0.14
Tomato	3	180–870	0.41 ± 0.17d	2	1.75 ± 0.85
Soybean	4	50–200	0.90 ± 0.38b	5	1.25 ± 0.62
Others	4	118–418	1.66 ± 0.88a	6	0.53 ± 0.08
† Peanut	1	207	0.22	1	1.0

† We excluded the peanut data from the independent t-test because it had just one N_2O measurement. Values are mean ± standard error. Different letters within the same column indicate significant differences at P < 0.05. Other vegetables for EF included amaranthus, brussels, lettuce, and turnip (n = 4), and amaranthus, brussels, turnip, pepper, and lettuce for BNE (n = 6).

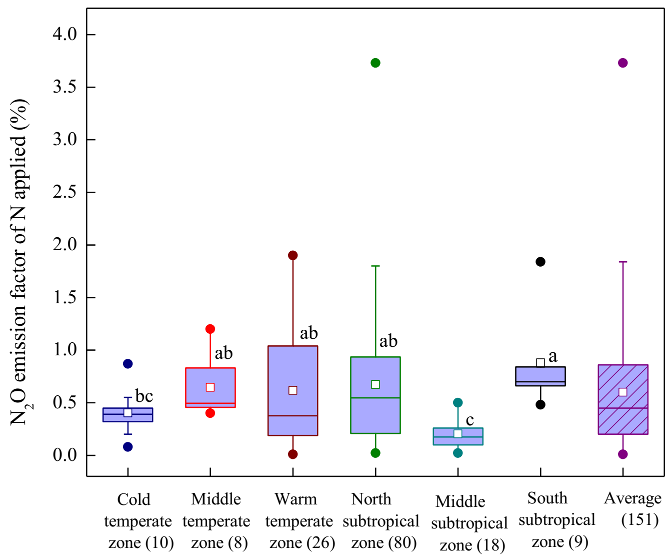

Fig. 2. Boxplots of N_2O emission factors of N applied across six climatic zones in China. The dots, the white square, and the rectangle denote the range, mean, and 95% confidence intervals, respectively. Values in parentheses indicate the number of measurements per group. Different lowercase letters indicate significant differences between the means at $P < 0.05$.

vegetables, at 90.0 Gg N yr^{-1}, followed by maize and wheat, at 66.8 and 49.3 Gg N yr^{-1}, respectively, with the lowest coming from rice (33.0 Gg N yr^{-1}); thus, their proportion of the total national N_2O emissions was 37.7% (vegetables), 27.9% (maize), 20.6% (wheat), and 13.8% (rice) (Table 4).

For the maize cropping system, the highest total N_2O emission was found in North China at 22.3 Gg N yr^{-1}, while South China had the lowest N_2O emission (1.59 Gg N yr^{-1}) (Table 4). Among provinces, the highest N_2O emissions came from Heilongjiang (8.92 Gg N yr^{-1}) and Jilin (6.70 Gg N yr^{-1}), respectively accounting for 13.4% and 10.0% of total national N_2O emissions from maize. Xizang (Tibet) had the lowest N_2O emission (0.005 Gg N yr^{-1}).

Under the rice cropping system, the highest N_2O emission occurred in East and Central China (15.1 Gg N yr^{-1}) and South China (8.75 Gg N yr^{-1}), respectively representing 45.5% and 26.4% of its total national emissions; the lowest N_2O emission was found in Northwest China (1.12%). For the provinces, the highest N_2O emissions were in Guangdong (4.76 Gg N yr^{-1}) and Jiangsu (4.44 Gg N yr^{-1}), corresponding to 14.4% and 13.4% of total national N_2O emissions, respectively, while just 0.001% was contributed by Beijing with the lowest N_2O emission (0.0002 Gg N yr^{-1}) (Table 4).

The highest N_2O emissions from wheat production were also found in North China (25.2 Gg N yr^{-1}) and East and Central China (15.5 Gg N yr^{-1}), respectively contributing 51.1% and 31.5% to the total national N_2O emissions for wheat, with South China contributing the least (0.03%). Among the provinces, the highest values were found in Henan (10.8 Gg N yr^{-1}) and Shandong (8.42 Gg N yr^{-1}), which constituted 21.9% and 17.1% of the total national emissions for wheat, respectively; Jilin had the lowest, contributing only 0.002% of the total national emissions.

For vegetables, the highest N_2O emissions were found in North China (26.7 Gg N yr^{-1}) and East and Central China (23.5 Gg N yr^{-1}), contributing 29.7% and 26.2%, respectively. Northeast China had the lowest emissions (4.68 Gg N yr^{-1}), contributing only 5.2% of the national total N_2O emissions under vegetable production. Comparing the provinces, the highest N_2O emission was in Shandong (10.6 Gg N yr^{-1}), contributing 11.8% to the total national emissions under vegetable cultivation, while Xizang (Tibet) had the lowest (0.08 Gg N yr^{-1}), contributing only 0.09%.

We found that the IPCC EF method for estimating N_2O emissions from China's rice, maize, wheat and vegetable fields was imprecise under certain conditions. The main factors influencing EF were soil pH and precipitation, with soil temperature and applied N less important. Using the regional or crop-specific method to calculate, these EFs evidently improved the confidence of our results. Fig. 5 shows the N_2O emissions from synthetic N fertilizer applied in croplands per province. The total annual N_2O emissions from maize, rice, wheat, and vegetable cropping systems in China, estimated using method 2 (crop-specific EFs) and method 3 (IPCC default EF), were 253 and 357 Gg N yr^{-1}, respectively (Table 3).

4. Discussion

4.1. Regional and crop-specific N_2O emission factors of N applied

The N_2O emission factor (EF) of synthetic N applied from croplands in China was estimated at 0.60%. This was much lower than the IPCC default value of 1.0% (IPCC, 2007), and those reported for Australia (0.9%), Africa (1.4%), Asia (1.1%), and the 1.3% measured in cornfields of the

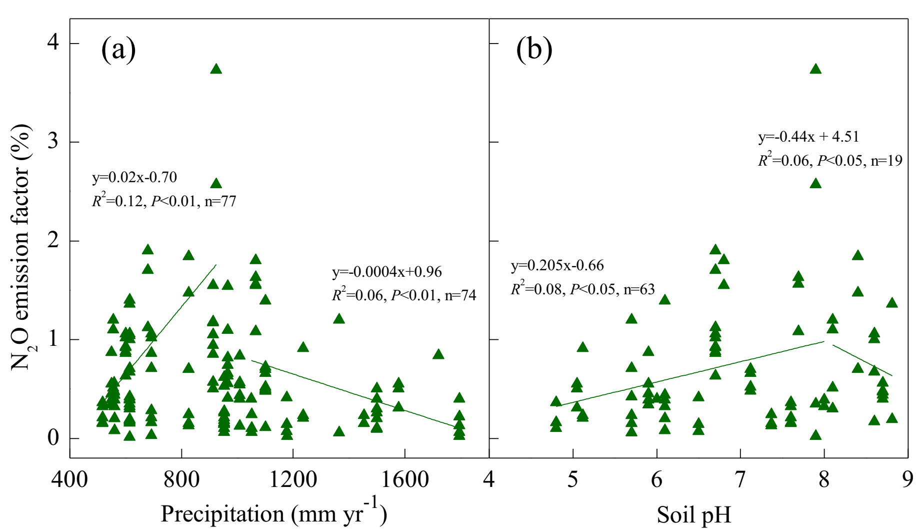

Fig. 3. Regression relationships between the N_2O emission factor of N applied and (a) precipitation, and (b) soil pH.

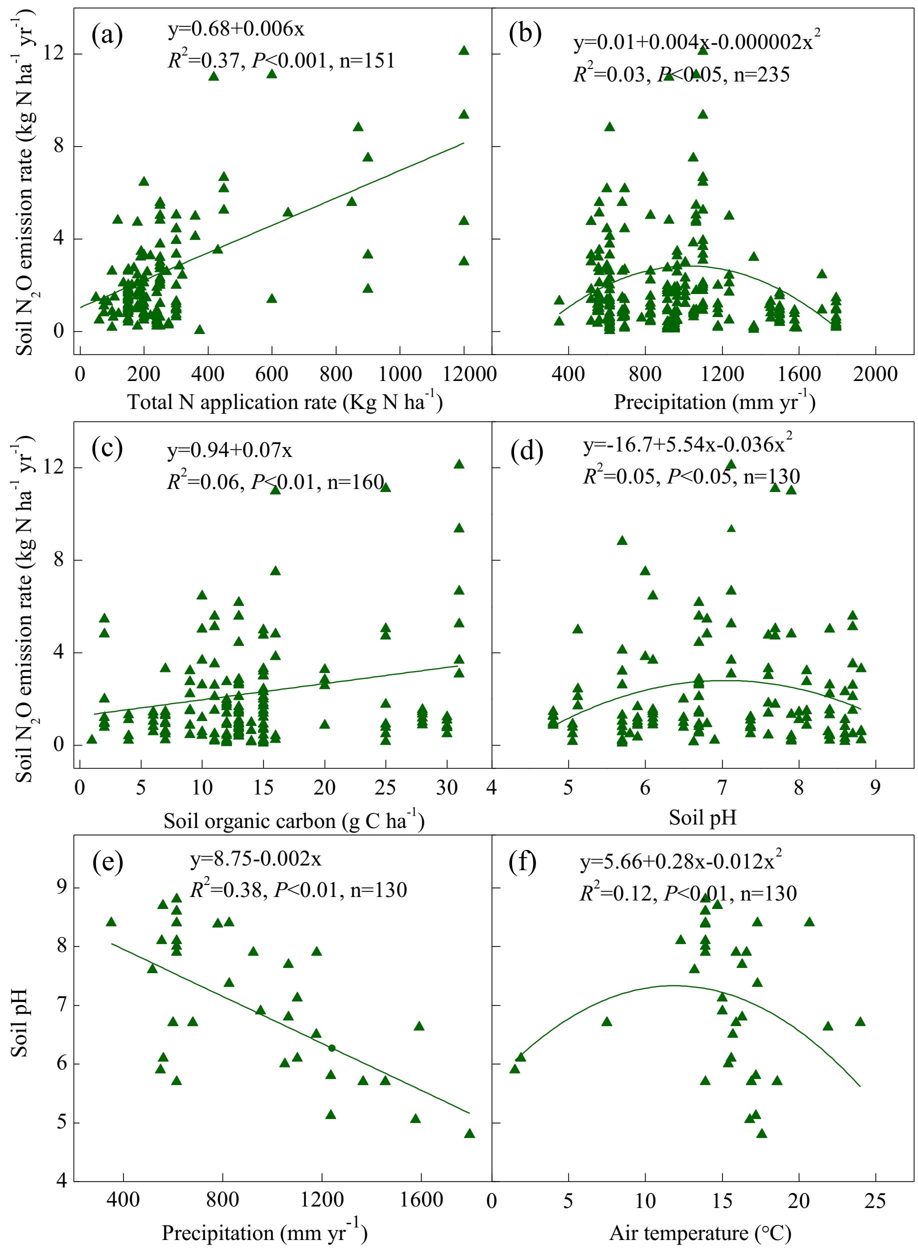

Fig. 4. Regression relationships between N_2O emission rate and (a) N application rates, (b) precipitation, and (c) soil organic carbon, and for soil pH against (d) precipitation and (f) air temperature.

USA (Griffis et al., 2013; Albanito et al., 2017), with a mean EF that was greater than the 0.02% of southern Spain (Menendez et al., 2008). Our EF was relatively similar to China's national average of 0.6% estimated previously by Xing (1998), but lower than 0.75% estimated for its paddy rice growth season (Zheng et al., 2004) and the 0.77% estimated for the uplands in China (Shepherd et al., 2015).

As expected, the EF differed considerably among the six climatic zone croplands in China, being significantly higher in the north and south subtropical zones than in the temperate zones. Consistence with this finding, Hirsch et al. (2006) found that 50–64% of global N_2O emissions originated from the tropics. China's high EF in its north and south subtropical zones probably arose because precipitation, soil pH, and air temperature there approached optimal values (pH, 5.5–7.3; air temperature, 25–40 °C) for nitrification and denitrification processes in soil (Granli and Bøckman, 1994; Curtin et al., 2012; Bouwman et al., 2002a; Stehfest and Bouwman, 2006). Earlier, Paul and Clark (1996) had reported that optimum pH values for nitrification and denitrification vary from 6.0 to 8.0, while their optimum temperature ranges from 20 °C to 35 °C (Vymazal, 1995; Barnard and Leadley, 2005). Generally, low temperatures tend to suppress the decomposition of soil organic carbon (SOC) and reduce the conversion rate of applied N to N_2O (Ding et al., 2013). China's north and southern subtropical zones receive heavy precipitation (Wang et al., 2017), which may induce peak N_2O emissions after fertilization (Ni et al., 2012; Chen et al., 2014), rendering the soil pH (5.05–8.4) in these zones more suitable for nitrification and denitrification, thereby contributing to high EFs (Hoben et al., 2011; Wang et al., 2018b).

Surprisingly, EF of synthetic N applied in China's cold temperate zone exceeded that in its middle subtropical zone. This was probably due to high emissions driven by denitrification during the freeze-thaw cycle (Brentrup et al., 2000; Kaiser and Ruser, 2000; Kim et al., 2012). For example, Chen et al. (2016) reported high N_2O emissions during

Table 3
Total soil N_2O emissions in different provinces estimated by using the regional EF, crop-specific EF, and IPCC default EF from China's main crop systems.

	Province	Climatic zone[a]	N_2O emissions (Gg N yr^{-1})		
			Regional EF	Crop-specific EF	IPCC EF value
North	Beijing	WTZ	0.56	0.58	0.79
	Tianjin	WTZ	1.02	1.07	1.43
	Hebei	WTZ	17.4	17.8	24.2
	Shanxi	MTZ	5.23	5.45	7.15
	Shandong	WTZ	25.3	25.7	35.4
	Henan	WTZ	25.5	25.9	35.9
Northeast	Liaoning	WTZ	8.94	9.77	13.4
	Jilin	MTZ	8.39	8.87	11.8
	Heilongjiang	MTZ	12.9	13.4	18.0
East and central	Shanghai	NSZ	1.21	0.95	1.42
	Jiangsu	NSZ	20.4	15.9	24.3
	Zhejiang	MSZ	2.01	4.29	6.32
	Anhui	NSZ	14.6	11.6	16.9
	Jiangxi	MSZ	2.72	5.12	7.85
	Hubei	NSZ	13.6	11.0	16.3
	Hunan	MSZ	5.29	11.0	16.9
South	Fujian	MSZ	2.37	5.31	7.79
	Guangdong	SSZ	11.9	8.87	13.4
	Guangxi	SSZ	9.21	7.21	10.2
	Hainan	ETZ	1.45	1.44	2.09
Southwest	Sichuan	MSZ	7.02	14.7	21.3
	Guizhou	MSZ	3.29	6.18	8.41
	Yunnan	SSZ	10.6	8.46	11.7
	Chongqing	MSZ	2.87	6.27	8.90
	Xizang	CPZ	0.14	0.14	0.18
Northwest	Neimongol	MTZ	7.71	8.10	10.4
	Shaanxi	WTZ	4.79	4.89	6.82
	Gansu	MTZ	5.75	5.99	7.96
	Qinghai	CPZ	0.32	0.31	0.40
	Ningxia	MTZ	1.49	1.56	2.11
	Xinjiang	MTZ	5.18	5.30	7.09
National total			239 ± 50.2	253 ± 53.1	357 ± 74.9

MTZ, middle temperate zone; WTZ, warm temperate zone; NSZ, north subtropical zone; MSZ, middle subtropical zone; SSZ, south subtropical zone; ETZ, edge tropical zone; CPZ, cold plateau zone.
[a] Classification based on Zheng et al. (2010) and Wang et al. (2017).

Table 4
Total N_2O emissions estimated from maize, rice, wheat, and vegetable cropping systems in different provinces of China by using the regional EFs.

	Province	N_2O emissions (Gg N yr^{-1})			
		Maize	Rice	Wheat	Vegetables
North	Beijing	0.17	0.0002	0.04	0.35
	Tianjin	0.31	0.02	0.17	0.52
	Hebei	5.66	0.09	4.64	7.04
	Shanxi	3.26	0.001	1.14	0.83
	Shandong	6.15	0.10	8.42	10.6
	Henan	6.76	0.59	10.8	7.40
Northeast	Liaoning	5.62	0.54	0.01	2.76
	Jilin	6.70	0.87	0.001	0.83
	Heilongjiang	8.92	2.82	0.07	1.09
East and central	Shanghai	0.01	0.22	0.14	0.84
	Jiangsu	1.35	4.44	6.90	7.74
	Zhejiang	0.07	0.40	0.10	1.44
	Anhui	2.01	2.96	5.82	3.80
	Jiangxi	0.03	1.55	0.01	1.13
	Hubei	1.88	3.53	2.52	5.64
	Hunan	0.34	1.98	0.03	2.94
South	Fujian	0.06	0.40	0.002	1.91
	Guangdong	0.47	4.76	0.002	6.69
	Guangxi	1.06	3.20	0.01	4.94
	Hainan	NC	0.39	NC	1.05
Southwest	Sichuan	1.40	0.98	1.06	3.58
	Guizhou	0.78	0.33	0.21	1.97
	Yunnan	3.28	2.24	0.74	4.31
	Chongqing	0.52	0.34	0.07	1.94
	Xizang	0.005	0.001	0.05	0.08
Northwest	Neimongol	5.79	0.08	0.95	0.90
	Shaanxi	0.24	0.11	2.05	2.40
	Gansu	1.59	0.01	1.13	3.03
	Qinghai	0.05	NC	0.11	0.16
	Ningxia	0.68	0.10	0.21	0.50
	Xinjiang	1.65	0.07	1.91	1.55
National total		66.8	33.0	49.3	90.0

NC: no cultivation.

the freeze-thaw period, which accounted for 20.1–49.4% of annual emissions in China's cold temperate zone when soil moisture was more than the 67–76% water-filled pore space (WFPS). By contrast, soils in the middle subtropical zone have a low pH (4.8) and SOC content (6.06 g C kg^{-1}), probably from low rates of organic matter incorporated into the soil. Long-term addition of compost alone, or in combination with inorganic fertilizers, increased SOC by 10–45% in the North China Plain (Ding et al., 2013), in line with the view that a high SOC in a relatively moist climate increases N_2O EF (Zhuang et al., 2012).

The highest EF arose from legume crops. This result is best explained by generally low N uptake and/or their ability to fix dinitrogen (N_2), the latter thus increasing soil N availability (Rochette and Janzen, 2005). According to Crutzen et al. (2008), the legume-rhizobium symbiosis could increase N_2O concentrations in the atmosphere, and N_2O emission rates from legume cultivated soils reportedly ranged from 1.9 to 2.9 kg N ha^{-1} yr^{-1}, averaging 2.4 kg N ha^{-1} yr^{-1} (Zhong et al., 2009). Nitrogen fixation estimates for various legume crops span 200 to 300 kg N ha^{-1} yr^{-1} (Peoples et al., 1995); as an example, estimates for pea, soybean, and cowpea are 23–300 kg N ha^{-1} yr^{-1} (Wani et al., 1995) or 65–335 kg N ha^{-1} yr^{-1} (Tate, 1995). Therefore, when soil mineral N availability exceeds plant uptake capacity, extremely high EF values are likely to occur (McSwiney and Robertson, 2005). N_2O emissions were detectable after roots and nodules decomposed during late growing stage or after harvesting (Rochette and Janzen, 2005; Yang and Cai, 2005; Zhong et al., 2011). Leguminous species with a covering crop had their N_2O emissions increased by 60% (Basche et al., 2014), because they released belowground N via biological N-fixation (Garland et al., 2014).

Interestingly, the EF from vegetable fields in China was 0.69%, hence lower than the 0.77% for vegetable fields in Japan (Greenhouse Gas Inventory Office of Japan, 2005), and close to the national EF for China's croplands. Although N_2O emission from vegetable fields was highest, its BNE was also the highest among cropping systems we investigated. Thus, it is likely that high BNE reduced N_2O emissions from an EF of N fertilizer applied in vegetable fields. Yet we also did not find an exponential relationship between EFs and the N application rate (Fig. 3c), unlike in other studies (e.g., Lu et al., 2006; Velthof and Mosquera, 2011). The higher EFs in vegetable than in cereal fields probably arise from increased nitrification and denitrification rates due to higher soil moisture, organic matter, and/or temperature (Lin et al., 2010), while the shallow root system and lower root density of vegetables diminishes their nutrient absorption capacity (Brumm and Schenk, 1992) promoting greater N_2O emissions (Wang et al., 2018a). Li et al. (2005) found that cereals, such as maize and wheat, can reduce soil nitrate accumulation, thereby reducing the potential risk of N leaching and pollution (Li et al., 2011). Our EF for cereals was similar to that reported from Germany (Jungkunst et al., 2006). That the maize's EF was greater than wheat's (Table 2) was primarily due to higher soil moisture and air temperature (Schils et al., 2008; Zhang and Han, 2008; Ni et al., 2012; Shepherd et al., 2015). Our results are in line with Ding et al. (2013), who reported an EF of 0.63% for a maize growing season that exceeded the wheat season's EF in China. The upland rice EF was higher, albeit not significantly than maize's EF, indicating that upland rice field is potential hotspots for N_2O emission, although its area is very low in China (1.77 Mha) (Grain and Oil Division of Planting Management Department of Ministry of Agriculture of China, 2003). This is probably because upland rice is mainly distributed in the

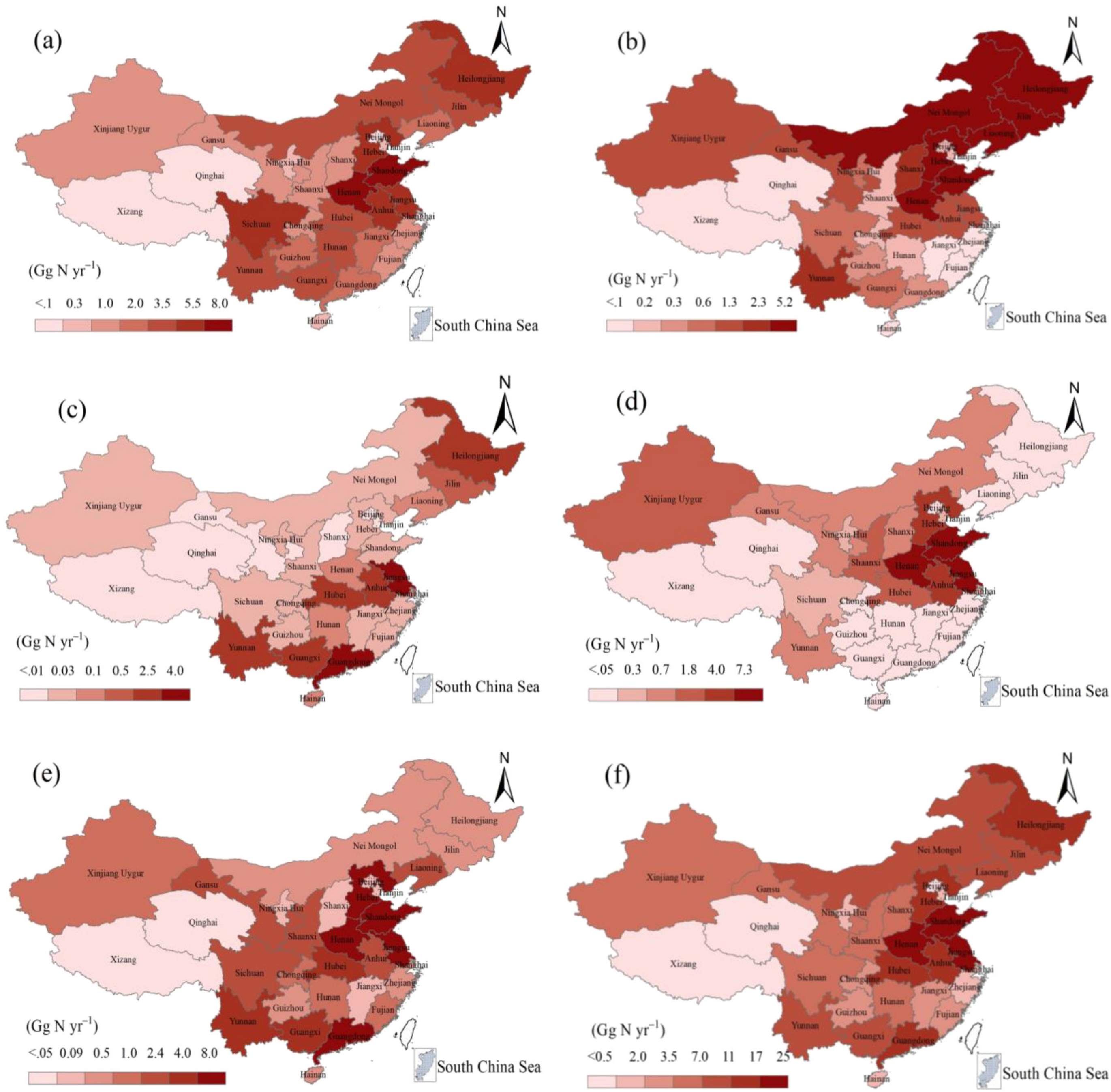

Fig. 5. (a) Total soil background N_2O emissions estimated by using the crop-specific background N_2O emission rate, and synthetic N applied N_2O emissions estimated by using the regional N_2O emission factor from fields of (b) maize, (c) rice, (d) wheat, and (e) vegetables based on 2016 cultivated (sowing) area in China (Gg N yr^{-1}) and (f) total soil N_2O emission (Gg N yr^{-1}).

subtropical mountain region with high precipitation and temperature (Defeng, 2000). Upland rice yields are comparably lower than those obtained from paddy rice, which attributed to small cultivated area (Defeng, 2000; Belder et al., 2005). In contrast, the paddy rice EF was significantly lower than those in maize and wheat fields (e.g. Gao et al., 2011; Zhou et al., 2014; Shepherd et al., 2015; H. Chen et al., 2015, J. Chen et al., 2015). In paddy field, flooding interspersed with midterm drainage and dry–wet alteration, flooding–midseason drainage–reflooding–final drainage, are common practices, which may create a favorable environment for short-term N_2O production and pulse emission (Zheng et al., 2000; Bouwman et al., 2002b; Zou et al., 2005a, 2005b; Wang et al., 2012). It is likely that rice EFs from paddies depends greatly on water management patterns (Zheng et al., 1999; Zou et al., 2003, 2007, 2009). There is still controversy about N_2O emissions associated with patterns of flooding or alternate wetting and drying (Kritee et al., 2018). Further studies are required in paddy fields with different water management patterns to improve the representativeness of rice-specific EF.

In this study, EF strongly depended on precipitation and soil pH but not the N application rate. Other reported associations between N application rate and EF in the literature vary: some studies fail to find any relationship between them (Zheng et al., 2004; Rashti et al., 2015; Wang et al., 2018a) while others suggest that EF increases with N fertilizer application rate (Velthof and Mosquera, 2011). For example, Lu et al.

(2006) observed that N input and precipitation were largely responsible for variability in the EFs they measured, while we found that the lowest EF (0.21%) was associated with the highest N rate applied (1200 kg N ha^{-1}) in cucumber fields while the highest EF (3.73%) was associated with N applied at 118 kg N ha^{-1} on amaranthus fields, an inverse relationship unheard of (Velthof and Mosquera, 2011; Signor et al., 2013). In particular, studies by Kim et al. (2013), Cayuela et al. (2017), and Wang et al. (2018a) all showed that EFs were relatively stable at different N fertilizer rates. Although EF and N rate were not significantly related, even within the same climatic zone, EF did sharply decrease when the N rate was above 400 kg N ha^{-1} in the fertilized crops. Although this finding is similar to those from agricultural systems in tropical and sub-tropical regions (Kim et al., 2013; Albanito et al., 2017), it should be interpreted cautiously because the N_2O EF expresses N_2O emitted relative to N input; hence, a decrease in EF with more N input in a C-limiting situation after peaking does not necessarily imply an absolute decrease in cumulative N_2O emissions (Kim et al., 2013).

The quadratic relationship between soil pH and EF suggested a neutral soil pH fostered the highest EF. Both fungi and bacteria preferred soil pH conditions near neutral to slightly alkaline (7.0–8.0) for nitrification and denitrification (Schmidt et al., 2011; H. Chen et al., 2015, J. Chen et al., 2015). In USA cornfields, a soil pH 6.3–6.8 had higher EFs that were lowered in soil pH 7.5 (Hoben et al., 2011; Shcherbak et al., 2014). As noted above, a soil pH of 6.0 to 8.0 is deemed optimal for nitrification and denitrification (Paul and Clark, 1996; Gieseke et al., 2006). Our non-linear relationships between EF and precipitation or soil pH, which have not been reported in other studies, indicate that for a given N fertilizer input, N_2O EFs tend to decrease more at pH >8.0 (Wang et al., 2018b), while precipitation and air temperature evidently explained some of the soil pH variation. Thus, it is likely that low precipitation and labile organic C reduced N_2O EF in soils with high organic C in China (Ni et al., 2012; Chen et al., 2014).

4.2. Estimation of total N_2O emissions from croplands

The total N_2O emission from synthetic N applied in vegetable, maize, rice and wheat cropping systems in China was estimated at 159 Gg N yr^{-1}. This accounted for 66.5% of the total N_2O emission during the vegetable, maize wheat and rice cropping seasons in 2016 which is within 64–75% obtained for the North China Plain (Ding et al., 2013). Compared with rice, vegetable crops produce greater N-induced N_2O emissions due to a higher N rate, similar to Europe and Northern America where non-rice crops had the highest N_2O emissions (Stehfest and Bouwman, 2006). Annual N fertilizer-induced N_2O emissions were much higher in Shandong and Henan provinces (warm temperate zone), mainly due to excessive use of fertilizer there coupled to a favorable soil pH (7.0–8.0) and soil moisture content >60% WFPS (He et al., 2007). Additionally, excess soil N surplus lowers plant N uptake efficiency, with the resulting soil residual N likely serving as substrates for additional N_2O emissions (Zebarth et al., 2008; Kim and Hernandez-Ramirez, 2010). Precipitation, irrigation, type of crops and higher N fertilizer inputs are all drivers stimulating N_2O emission in warm temperate zones (Garland et al., 2011). For example, Philibert et al. (2013) identified N fertilization, type of crop and experimental duration as the three most important factors of N_2O emissions using Random Forest. The lower emission, of 16.6 Gg N yr^{-1} from Northwest China (or 10.4% of the national N fertilizer induced N_2O emissions) is best explained by this region's low temperatures and precipitation coupled to a low N input rate, averaging 245 kg N ha^{-1} (Sun et al., 2016). This result is consistent with highly heterogeneous N_2O emissions observed across crops and regions by Gerber et al. (2016).

When derived from the regional and crop-specific methods, total national N_2O emissions from fertilized croplands for maize, rice, wheat and vegetable were 239 and 253 Gg N yr^{-1}, respectively. However, both estimates were lower than those calculated using the IPCC EF under IPCC Tier 1 guidelines (Table 3), corresponding to a non-trivial difference of 118 Gg N yr^{-1}, equivalent to a 33% increase. Recently, Wang et al. (2018b) suggested that adopting the IPCC Tier 1 method overestimated N_2O emissions by 38–46% in China. In contrast, these were underestimated in the USA's corn belt, at only 230 Gg N yr^{-1} following the IPCC methodology versus global top-down EFs of 420 Gg N yr^{-1} of N_2O emission when measured directly from cropland (Griffis et al., 2013). Philibert et al. (2012) observed that the range of uncertainty for N_2O emissions estimated with the IPCC Tier 1 default EF method was larger than that evaluated with the eight selected linear and nonlinear models. Similarly, Verhoeven et al. (2017) found that actual emission estimates for California's cropland deviated from those based on the IPCC method. Interestingly, the IPCC-based estimates substantially differ from those using our crop-specific method, the latter may have provided a more reasonable estimate for the total N_2O emission, since EFs can vary greatly among crop types (Shepherd et al., 2015). Zheng et al. (2004) reported that although some crop EFs were lower and others were higher than the IPCC default value, the total estimate was slightly lower, but still similar to that obtained via the IPCC method.

Taking the regional EFs into account greatly improved N_2O emission estimates from croplands in China when compared with only mean national or IPCC EFs; however, our estimate might harbor uncertainties. The size of the confidence interval is smaller for methods 1 and 2 to that defined by the IPCC-Tier 1 default method, particularly for average N application rate commonly used in China of 263 kg N ha^{-1}, with an uncertainty range of 235–291 kg N ha^{-1} ($n = 121$) (Table S3). The lower and upper limit of the IPCC range gives an estimated N_2O emission of 0.53–7.90 kg N_2O-N ha^{-1} yr^{-1} for an average N dose of 263 kg N ha^{-1}, whereas the lower and upper limit of methods 1 and 2 ranges from 0.01 to 5.04 kg N_2O-N ha^{-1} yr^{-1} and 0.09–5.55 kg N_2O-N ha^{-1} yr^{-1}, respectively (Table S6, Fig. S1). The rate of increase of N_2O emissions estimated with the three models with the amount of N applied was greater with IPCC model than with the other two models for maize, rice, wheat and vegetables. This result is consistent with the findings of Philibert et al. (2012), suggesting that the IPCC EF value was overestimated particularly for N application rates below 300 kg N ha^{-1}, as commonly used in agriculture. Our uncertainties are within the range of 0.01–15.6 kg N_2O-N ha^{-1} yr^{-1} calculated for Asian croplands (Philibert et al., 2012). Another set of uncertainties existed between experimental variability of N_2O emission. Firstly, some uncertainty may arise from the sampling frequency of the N_2O flux measurements, since N_2O emissions from cropland are often emitted in pulses (Smith and Dobbie, 2001; Zheng et al., 2004; Xiong et al., 2006; Ju et al., 2011; Philibert et al., 2012; Chen et al., 2016). Secondly, N_2O measurements were not available for the cold plateau and marginal tropical zones; hence, the national average EF was used for estimation. This could cause huge uncertainties in our regional estimate of N_2O emissions due to limited sample sizes in specific climate regions. Hence, more field measurements are needed to improve the accuracy of N_2O emission estimates using the regional EFs. Thirdly, sowing area used from the China Statistical Year Book (2016) is subject to land use change (Liu et al., 2006). Meanwhile, part of paddy fields especially in the North subtropical zone was fallow rather than cultivated with wheat in winter as usual. Fourthly, there were differences in type and amount of synthetic N fertilizer applied in different provinces among four different crops (Roelandt et al., 2005; Philibert et al., 2012; Zou et al., 2009; Gao et al., 2011). Fifthly, a single rice-specific EF (rather than based on different water management regimes) would lead to overestimation of N_2O emissions (Zou et al., 2009). Unfortunately, we found only limited published data for legumes, so emission from legume cropping system with high N_2O EF was not included in estimate, likely increasing uncertainties of estimation in this study. Our current estimations could be improved when a substantial number of

field measurements with detailed attributes are available under different climatic conditions in the future.

5. Conclusions

Overall, our results show that N_2O emission factors (EFs) in croplands in China averaged 0.60%. But there were significant differences between different climatic zones across the country, with the highest EF for upland fields in the north subtropical zone (0.93%) and the lowest for paddy field in the middle subtropical zone (0.20%). Precipitation and soil pH were among the factors explaining regional disparities in EFs, while synthetic N fertilizer input and precipitation were key factors promoting N_2O emissions. Total N_2O emission from croplands in China, calculated using our regional EF method was substantially (33%) lower than that derived from the IPCC's default EF calculation. We conclude that using a regional (method 1) or crop-specific EFs (method 2) is more accurate for estimating N_2O emission in China's cropland China than relying on the IPCC EF default value (method 3), so these should be included to ensure accurate N_2O inventories. However, as more field measurements from various cropping systems under different climatic zones become abundant, the integration of regional and crop-specific EFs (methods 1 and 2) could prove even more accurate for estimating N_2O emissions than using the regional EFs (method 1) alone.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2019.03.142.

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